# Airborne measurements of submicron aerosols across the coastline at Bhubaneswar during ICARB

P MURUGAVEL, V GOPALAKRISHNAN, VIMLESH PANT and A K KAMRA

Indian Institute of Tropical Meteorology, Dr. Homi Bhabha Road, NCL Post, Pune 411 008, India.

Airborne measurements of the number concentration and size distribution of aerosols from 13 to 700 nm diameter have been made at four vertical levels across a coastline at Bhubaneswar (20°25'N, 85°83'E) during the Integrated Campaign for Aerosols, gases and Radiation Budget (ICARB) programme conducted in March–April 2006. The measurements made during the constant-level flights at 0.5, 1, 2 and 3 km altitude levels extend ~100 km over land and ~150 km over ocean. Aerosol number concentrations vary from 2200 to 4500 cm<sup>-3</sup> at 0.5 km level but are almost constant at ~ 6000 cm<sup>-3</sup> and ~ 800 cm<sup>-3</sup> at 2 and 3 km levels, respectively. At 1 km level, aerosol number concentration shows a peak of 18,070 cm<sup>-3</sup> around the coastline. Most of the aerosol size distribution curves at 0.5 km and 1 km levels. However, at the peak at 1 km level, number concentration has a bimodal distribution with an additional maximum appearing in nucleation mode. It is proposed that this maxima in nucleation mode at 1 km level may be due to the formation and transport of new particles from coastal regions.

# 1. Introduction

Physical and chemical properties of the marine and continental aerosols are significantly different since the sources and sinks for the two types of aerosols widely differ from each other (see e.g., Fitzgerald 1991; Jaenicke 1993). Therefore, properties of the aerosols over coastal regions where marine and continental aerosols intermix with each other, are quite complex. Further, complicating the situation is the fact that coastal regions have prominent sources such as the breaking waves in the surf region and the gas-to-particle conversion processes for production of both primary and secondary marine aerosols (Blanchard and Woodcock 1980; O'Dowd et al 1997, 1998, 1999, 2007; Clarke et al 1998). The aerosols produced in coastal regions are transported by various land-to-sea and sea-to-land transport processes which cause them to disperse in the atmosphere, both horizontally and vertically. These transport processes range from the large-scale atmospheric circulation to

the regional and local transport processes such as sea- and land-breezes and convective motions of the atmosphere. Further, the vertical stability of the lower atmosphere strongly affects vertical distribution of aerosols in these regions. As a result of a variety of these production and dispersal processes, the atmospheric aerosols over coastal regions undergo large temporal and spatial variations.

In order to explore the nature of aerosols in coastal regions, the New Particle Formation and Fate in the Coastal Environment (PARFORCE) experiment was conducted in which an aircraft was used as platform to map the aerosol distributions over Mace Head on Irish west coast (O'Dowd *et al* 1998, 1999; O'Dowd 2002). In this experiment, the coastal plume was observed to advect upto 250 km offshore and the nucleation mode was observed to grow in sizes >40 nm in ~3 hours. The experiment confirmed that the source of coastal nucleation events is indeed the tidal zone around the coastline and these aerosols are convected up in the

Keywords. Atmospheric aerosols; coastal aerosols; new particle formation; nucleation mode; aerosols; atmospheric sciences.

P Muruqavel et al



Figure 1. Flight tracks of the aircraft at four different levels. The numbers along the tracks show the mean time of taking an air sample for determination of size distribution.

atmosphere. Such coastal nucleation events have been reported from several other coastal locations (Aitken 1897; Paugam 1975; Bigg and Turvey 1978; Grenfell *et al* 1999; O'Dowd *et al* 1996, 1997).

In this paper, we present results of our airborne measurements of the concentration and size distribution of submicron aerosols made at four vertical levels during a flight on March 25, 2006 across the coastline at Bhubaneswar (20°25′N, 85°83′E), India during the Integrated Campaign for Aerosols, gases and Radiation Budget (ICARB). The flight track in the east–west direction extended partly over land and partly over ocean.

# 2. Instrumentation

A Scanning Mobility Particle Sizer (SMPS), Model 3080, of TSI, installed onboard a beachcraft aircraft is used to make measurements of the number concentration and size distribution of aerosol particles in the size range of 13 to 700 nm. The air sample is taken through an air inlet fixed at the belly of the aircraft and is located  $\sim 2 \text{ m}$  away from the nose of the aircraft. The inlet consists of a metal pipe of 5 mm outer diameter and 4 mm internal diameter extending 10 cm from the body of the aircraft and is smoothly bent to face the airflow. The ambient air enters the inlet tube and is fed to the instrument through a Teflon tube of 4 mm internal diameter. The instrument is kept close to the inlet so as to minimize the transmission losses of aerosol particles inside the tube.

Aerosol size distributions are measured along the flight track during the level flights only. Since the time period required for taking one size distribution is 3 minutes, 7–12 size-distributions are taken at each level. The SMPS is operated in low-flow mode which requires a total airflow rate of 3 lpm and aerosol flow rate of 0.3 lpm. The flow rates are adjusted at each level so as to keep a constant volume flow of 3 lpm. The air samples taken during the ascents and descents of the aircraft are not considered.

The airborne sampling of aerosols is prone to errors that may be introduced by the change in flow rate through the instrument due to changes in pressure and temperature with altitude. However, these errors may not exceed  $\pm 10\%$  as SMPS has been designed to work upto 3000 m altitude with  $\pm 10\%$  accuracy and our measurements have been made below 3000 m. Further, there may be some uncertainty in the measured aerosol number concentration due to counting statistics (de Reus *et al* 2001). However, such an error is limited only to 3–4% for particles <40 nm diameter and <2% for particles of diameter >40 nm. To minimize diffusion losses, sampling was done at a distance of only about 2 m from the air inlet. The diffusion losses are ~7% for 13 nm diameter particles and <1% for particles of 540 nm diameter (Baron and Willeke 2001). As the inlet and the tube taking the air sample to the instrument were designed to have no sharp or right-angled bends, the losses introduced due to such bends are negligible.

# 3. Flight track

Measurements made on a flight on April 25, 2006 during the ICARB are presented here. The flight started from Bhubaneswar at 0330 UT. The aircraft climbed to the 500 m level and then proceeded eastwards, crossed the coastline and then continued its flight over the Bay of Bengal. In total, the track at this level covered  $\sim 100 \,\mathrm{km}$  over land and  $\sim 150 \,\mathrm{km}$  over sea. The aircraft then climbed to 1000 m and flew back towards Bhubaneswar. Almost similar tracks were made from sea to land at 1000 m, from land to sea at 2000 m and again from sea to land at 3000 m level, above each other. Figure 1 shows the flight tracks at four different levels. The labels along the tracks show the mean time of taking an air sample for determination of size distribution and the arrows indicate the direction of flight. The thick line shows Indian landmass to the left and the Bay of Bengal to the right.

# 4. Vertical stability of the lower atmosphere

Vertical profiles of potential temperature, relative humidity and mixing ratio calculated from radiosonde flights made at 00 UT at Bhubaneswar on March 25, 2006 are plotted in figure 2. Over a highly stable layer of 500 m thickness, there is a ground inversion at 500 m. The atmosphere continues to be stable up to  $\sim 1800 \,\mathrm{m}$ . From 1800 to  $\sim 2200 \,\mathrm{m}$  the atmosphere is locally unstable, perhaps due to local advection, and is capped by an inversion layer. Above  $\sim 2200 \,\mathrm{m}$ , the atmosphere is again highly stable. Mixing ratio of  $20 \text{ gm kg}^{-1}$  and relative humidity of >90% at the surface indicate fog conditions but the atmosphere soon dries up at  $\sim 800 \,\mathrm{m}$  level. Slight increase in relative humidity between 1500 m and 2900 m is due to local advection. Southerly surface winds of  $\sim 5\,\mathrm{m~s^{-1}}$  changed to westerlies at 1500 m and then intensified to northerlies of jet speed of  $\sim 10 \,\mathrm{ms}^{-1}$  at 3000 m level.



Figure 2. Profiles of air temperature, dewpoint temperature, potential temperature, relative humidity, mixing-ratio calculated from the radio sonde flights made at Bhubaneswar at 00 UT on March 25, 2006.



Figure 3. The 5 days back trajectories at different levels at 0400 UT on 25 March, 2006 obtained from NOAA-HYSPLIT model.

#### 5. Back trajectories

The 5 days back trajectories calculated from the HYSPLIT model for the airmasses arriving at different levels over Bhubaneswar are shown in figure 3. The places from where the air parcels



Figure 4. Total number concentration of aerosols along the flight track at different levels. Vertical continuous lines divide the four segments of flight as shown in figure 1.

arrive at these levels, widely differ. While at 500 m level, the air parcel is being advected from west Pakistan, the air parcels at 1000 m and 2000 m are being advected from the desert regions of Iran and Saudi Arabia, respectively. The air parcel at 3000 m is being advected from the central Arabian Sea. It travels west but then recurves to pass over the eastern coastline of Arabia and the Arabian Sea and enters the east coast of India to pass over the Thar Desert.

The air parcels reaching at 2000 and 3000 m levels have long journeys of about one-and-a-half and three-and-a-half days, respectively, over water. Further, the air parcels reaching from 1000 to 3000 m levels descend from 4 to 5 km levels. One thing common in trajectories at all levels is that in the last 2–3 days, all air parcels, travel from northern India to eastern coast of India a little south of the Indo-Gangetic Plains.

#### 6. Observations

Total aerosol number concentrations integrated from each aerosol size distribution measured at four levels along the flight track, are plotted in figure 4. Vertical continuous lines in figure 4 separate the segments of the flight track at different altitudes and vertical dashed lines in each segment show the position of coastline. X axis shows the mean time of

taking an air sample for measurement of the aerosol size distribution. At 500 m level, variations in total number concentration which varies between 2200 and  $4500 \,\mathrm{cm}^{-3}$  do not show a definite trend with distance until one reaches close to  $\sim 40 \,\mathrm{km}$  from the coastline from where the concentration starts decreasing as one approaches the coastline and continues to decrease over the ocean as the distance from the coastline increases. This observation is in conformity with the surface observations of the land-to-ocean transport of aerosols made on the ship cruises as they sail away from the coastline (Hoppel and Frick 1990; Deshpande and Kamra 1995, 2002). Our observations show that such a land-to-ocean dispersal of aerosols extends at least up to an altitude of  $0.5 \,\mathrm{km}$  in a stable atmosphere. When the aircraft climbs to 1000 m level over sea and starts its flight towards land, total aerosol concentration first increases to  $\sim 4500 \,\mathrm{cm^{-3}}$  and remains almost constant at this level for the first 15 minutes when the aircraft is  $\sim 75 \,\mathrm{km}$  away from the coastline. Then total aerosol concentration sharply increases and reaches a peak value of  $18070 \,\mathrm{cm}^{-3}$  about  $25 \,\mathrm{km}$  away from the coastline, on landside. The concentration then sharply decreases to  $\sim 8000 \,\mathrm{cm}^{-3}$  and remains almost constant. Aerosol number concentration is highest at this level as compared to that at other levels and peaks nearly over the coastline. At 2000 m level, the number concentration decreases to  $\sim 6000 \,\mathrm{cm^{-3}}$ 



Figure 5. Vertical profiles of total number concentration of aerosols over ocean and over land.

and remains nearly constant as one moves from land to ocean; it does not show any peak at the coastline as at 1000 m level. Aerosol number concentration further falls to  $<1000 \,\mathrm{cm^{-3}}$  at 3000 m level and remains almost constant at this level both over the ocean and land.

Figure 5 shows the vertical profiles of total aerosol number concentration averaged separately for all measurements taken vertically over land and over ocean. Total number concentrations are higher over land than that over ocean throughout the depth of boundary layer and became almost equal above  $\sim 2000 \,\mathrm{m}$ , i.e., above inversion layer.

Figures 6(a) and 6(b) show all the aerosol size distributions observed at each level over land and ocean, respectively. Nearly all the size distributions over land and over sea are monomodal with a maximum at 110 nm diameter at 500 m and 1000 m levels that shifts to a lower value of 70 nm diameter at 2000 m and 3000 m levels. However, height of the maxima is maximum mostly over or near coastline. Figure 7 shows a three-dimensional view of the aerosol size distributions at each of the four levels. An important feature of the size spectra plotted in figures 6 (a and b), and figure 7 is the observation of higher concentrations of Aitken particles of  $< 50 \,\mathrm{nm}$  diameter particles in the size distribution curves measured over or near coastline at and above 1000 m. Some of the curves measured near coastline at 1000 m show even a second prominent maximum for Aitken particles. We assign the peak observed at 4.52.29 UT in figure 4 to the newly formed particles in the low tide regions of coastline and the almost constant but enhanced values of number concentration before and after the peak to the changes in altitude of measurements. Further, proportional contribution of Aitken particles to

total aerosol concentration increases very much at 3000 m.

# 7. Discussion

Large scale subsidence in the area of measurement during this season and the morning period of making measurements suggest that the measurements have been made in a stratified atmosphere. The vertical profiles of thermodynamic parameters, plotted in figure 2 also support it. Under such conditions, horizontal advection of airmasses, as shown by air trajectories in figure 3, is expected to play a prominent role in vertical distribution of aerosols. The airmasses horizontally advected from the desert regions are most probably responsible for comparatively higher total aerosol number concentrations at 1000 m and 2000 m levels. In the absence of any vertical transfer above inversion, very low values of total aerosol concentrations observed at 3000 m level are most likely associated with the air sample horizontally advected from the central Arabian Sea region. The observation of a decrease in concentrations, both above and below the layer of maximum concentration at 1000 m, also supports that the particles in this layer are being advected horizontally. Such layers of high aerosol concentration have been earlier reported to be observed in stratified atmosphere (Clarke and Kupustin 2001; Roberts et al 2006).

Formation of new particles in the low tide conditions in coastal regions due to emission of DMS by the marine biotic life has been proposed as a strong source for particles in coastal regions. These particles are only a few nanometer in size at the time of formation but can grow to Aitken particle size range in at few hours. O'Dowd (2002) has observed that the nucleation mode particles generated in tidal zone around the coastline at Mace Head can be mixed up to more than 1000 m in the vertical, inspite of slightly stable meteorological conditions of the lower atmosphere. The Aitken particles observed over the coastline region at 1000 m altitude or above in our measurements are most likely formed in the coastal regions. At the time of formation, and at 500 m level, such particles may be only a few nanometer in size and thus may not be counted with our instrument. But they may grow to measurable sizes by the time they are transported to 1000 m. The fact that size distributions at both, 500 and 1000 m levels, show their maxima at the same diameter also indicates extension of the same or similar airmass at these levels. Modeling studies of Pirjola et al (2000) show that the enhancement of CCN concentrations during their advection may be due to both, the condensation of sulphuric acid and biogenic vapour.



Figure 6. Aerosol number size distributions observed at different times (a) over land and (b) over ocean at different altitudes. Note the change in the vertical scale at 3000 m.



Figure 7. The three-dimensional views of aerosol size distribution obtained at different levels. The arrow on the x axis indicates the position of coastline. Note the change in the vertical scale at 3000 m.

#### 8. Conclusions

Our measurements show that the land-to-ocean dispersion of aerosols in stable atmosphere may extend upto 0.5 km altitude. Further, in agreement with O'Dowd's (2002) observations, our results show that the newly-formed particles in coastal zones may be convected upto 1 km in altitude even in stable meteorological conditions in the lower atmosphere. Moreover, under large-scale subsidence and stable atmospheric conditions, horizontal advection from Arabia and the Arabian Sea may play a dominant role in vertical distribution of aerosols over the coastline near Bhubaneswar.

# Acknowledgements

The authors are thankful to the organizers of the ICRAB of ISRO-GBP for their participation in the experiment and to Dr Kalyanaraman and Mr Raghu Venkataraman of National Remote Sensing Agency, Hyderabad, for providing the aircraft. They also gratefully acknowledge the NOAA Air Research Laboratory (ARL) for the provision of the HYSPLIT transport and dispersion model and/or READY website (http://www.arl.noaa.gov/ready.html) used in this publication.

#### References

- Aitken J A 1897 On some nuclei of cloudy condensation; Tran. R. Soc. Edin., XXXIX.
- Baron P A and Willeke K 2001 Aerosol Measurement: Principle, Techniques and Applications, John Wiley & Sons, 56–60.
- Bigg E K and Turvey D E 1978 Sources of natural atmospheric particles over Australia; Atmos. Environ. 12 1642–1655.
- Blanchard D C and Woodcock A H 1980 Production, concentration, and vertical distribution of the sea salt aerosols; Ann. NY Acad. Sci. **353** 186–200.
- Clarke A D, Warner V J, Eisele F, Mauldin R L, Tanner T and Litchy M 1998 Particle production in the remote marine atmosphere: cloud outflow and subsidence during ACE-1; J. Geophys. Res. 103 16,397–16,409.
- Clarke A D and Kapustin V N 2001 Aerosol climatology of the pacific: Production, transport and evolution of natural and anthropogenic particles, 8<sup>th</sup> European symposium on the Physico-chemical behaviour of atmospheric pollutants, Torino.
- de Reus M R, Krejci, Williams J, Fischer H, Scheele R and Storm J 2001 Vertical and horizontal distributions of the aerosol number concentration and size distribution over the northern Indian Ocean; J. Geophys. Res. 106 28,629–28,641.

- Deshpande C G and Kamra A K 1995 Extension of atmospheric aerosols over ocean around peninsular India in the southwest monsoon season; *J. Aerosol Sci.* **26** 1169–1174.
- Deshpande C G and Kamra A K 2002 Aerosol size distribution in the north and south Indian Ocean during northeast monsoon season; Atmos. Res. 65 51–76.
- Fitzgerald J W 1991 Marine aerosols: a review; Atmos. Environ. 25A(3/4) 533–545.
- Grenfell J L, Harrison R M, Allen A G, Shi J P, Penkett S A, O'Dowd C D, Smith M H, Hill M K, Robertson L, Hewitt C N, Davison B, Lewis A C, Creasy D J, Heard D E, Hebestreit K, Alicke B and James J 1999 An analysis of rapid increase in condensation nuclei concentrations at a remote coastal site in western Ireland; J. Geophys. Res. 104 13,771–13,780.
- Hoppel W A and Frick G M 1990 Submicron aerosol size distribution measured over the tropical and south pacific; *Atmos. Environ.* **24A** 645–659.
- Jaenicke R 1993 Tropospheric aerosols; In: Aerosol-Cloud-Climate Interactions, P V Hobbs (ed.) Academic Press, Inc., pp 1–31.
- O'Dowd C D 2002 On the spatial extent and evolution of coastal aerosol plumes; J. Geophys. Res. 107(D19) 8105, doi:10.1029/2001JD000422.
- O'Dowd C D, Smith M H, Lowe J A, Davison B M, Hewitt C N and Harrison R M 1996 New particle formation in marine environment, Proc. of 14<sup>th</sup> Int. Conf. on Nucleation and Atmospheric aerosols; Kulmala M and Wangner P (eds) (New York: Pergamon Press) pp. 925–929.

- O'Dowd C D, Davison B, Lowe J A, Smith M H, Harrison R M and Hewitt C N 1997 Biogenic sulphur emissions and inferred sulphate CCN concentrations in and around Antarctica; J. Geophys. Res. 102 12,839–12,854.
- O'Dowd C D, Geever M, Hill M K, Smith M H and Jennings S G 1998 New particle formation: Nucleation rates and spatial scales in the clean marine coastal environment; *Geophys. Res. Letts* **25** 1661–1664.
- O'Dowd C D, McFiggens G M, Pirjola L, Creasey D J, Hoell C, Smith M H, Allan B J, Plane J M C, Heard D E, Lee J D, Pilling M J and Kulmala M 1999 On the photochemical production of new particles in the coastal boundary layer; *Geophys. Res. Letts* **26** 1707–1710.
- O'Dowd C D, Yoon Y J, Junkerman W, Aalto P, Kulmala M, Lihavainen H and Viisanen Y 2007 Airborne measurements of nucleation mode particles I: coastal nucleation and growth rates; *Atmos. Chem. Phys.* 7 1491–1501.
- Pirjola L, O'Dowd C D, Brooks I M and Kulmula M 2000 Can new particle formation occur in the clean marine boundary layer? J. Geophys. Res. 105 26,531–26,546.
- Paugam J Y 1975 Formation de noyaux Aitken dans l'air au-dessus de la zone littorale; J. de Recherches Atmospheriques 9 67–75.
- Roberts G, Mauger G, Hadley O and Ramanathan V 2006 North American and Asian aerosols over the eastern Pacific Ocean and their role in regulating cloud condensation nuclei; J. Geophys. Res. 111 D13205, doi:10.1029/2005JD006661.

MS received 31 July 2007; revised 18 September 2007; accepted 10 October 2007